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**A Method for Manufacturing a Polyester Resin for Use in Food
Containers**

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For: Eastman Chemical Company

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

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| (73) Patent Holder: | Kim Jun-eung, Sonkyong Industry Co., Ltd. Bldg. 600, Jongja 1-dong, Jangan-ku, Suwon-shi, Kyonggi-do | |
| (72) Inventors: | Kim Jong-rang Bldg. 128, No. 201, Jugong Apts., Chonchon-dong, Jangan-ku, Suwon-shi, Kyonggi-do Park Hui-jong Bldg. 101, No. 510, Hanguk Apts., Metan 2-dong, Paldal-ku, Suwon- shi, Kyonggi-do | |
| (74) Agent: | Ho Sang-sun | |
| <i>Examiner: Lee Jae-eung (Pamphlet Publication No. 5016)</i> | | |
| (54) [Title of the Invention]: | A Method for Manufacturing a Polyester Resin for Use in Food Containers | |

Abstract

No abstract.

Specification

[Title of the Invention]

A method for manufacturing a polyester resin for use in food containers.

[Detailed Description of the Invention]

The invention relates to a method for manufacturing a polyester resin for use in food containers, and more specifically, an economical method for the manufacture, without using solid-state polymerization, of a polyester resin suitable for use in containers for carbonated beverages, bottled water, and fruit drinks, etc., in which, during manufacturing of the polyester polymer, an inert gas is blown into the melt to form prepolymer pellets containing air bubbles,

said pellets are heated, and a vacuum is simultaneously applied in order to manufacture polymer pellets, after which crystallization and drying are carried out.

There is currently a dramatic increasing trend in the demand for polyester resins for use in food containers, and in order to meet this demand, melt polymerization followed by solid-state polymerization has conventionally been used in manufacturing these polyester resins, particularly polyethylene terephthalate (abbreviated in the following as PET), in order to impart various physical properties to the resin.

In manufacturing of PET in particular, the advantages of solid-state polymerization of polyester resins include increased strength due to the formation of high-molecular-weight compounds and the elimination of acetaldehyde, which adversely affects the taste of food. With the recent development of melt polymerization methods, it has become possible to easily obtain polymers having a sufficient molecular weight for use in containers by means of processes in which melt polymerization is used, without the solid-state polymerization process, but it has not yet been possible to solve the problem of impaired taste.

The inventors of the invention therefore conducted research on methods for economically manufacturing polyester resins for use in food containers without carrying out solid-state polymerization, developing a process in which an inert gas is blown in during the melt polymerization process in order to manufacture polymer pellets that are then crystallized and dried, thus arriving at the invention.

The object of the invention is to provide a new method for manufacturing polyester resin for use in food containers without using the solid-state polymerization process.

The following is a detailed description of the invention.

The invention relates to a method for manufacturing a polyester resin for use in food containers in which, during manufacturing of the polyester polymer, an inert gas is blown into the melt to form polyester prepolymer pellets containing air bubbles, said pellets are heated under a vacuum to obtain popcorn-type polymer pellets, after which crystallization and drying are carried out.

The following is an explanation of the invention in greater detail.

The invention relates to a process in which a polyester resin for use in food containers is manufactured by means of the method of blowing an inert gas into the melt polymer during the melt polymerization process. During this process, the inert gas is blown in from the bottom of the polymerization reactor during melt polymerization in order to form polyester prepolymer pellets containing air bubbles, the pellets are crystallized by increasing the temperature from room temperature under a vacuum, causing the pellet-shaped chips to break up and take on the appearance of popcorn, these popcorn-like polyester polymers are crystallized, and acetaldehyde is removed.

The melt polymerization of the polyester of the invention is carried out according to a method commonly known to the one skilled in the art, and the reaction temperature, catalysts, catalyst content, and stabilizers used are all well-known in the literature and in the field of polyester prepolymer manufacturing.

The polyester prepolymer used in the invention is a commonly-known polymer mainly containing PET or 80 mol % or more of ethylene terephthalate structural units, and polyesters allowing solid-state polymerization, such as polyethylene isophthalate, polycyclohexane diethylterephthalate, and polyethylene naphthalate may also be used.

Moreover, the polyester prepolymer may also contain dicarboxylic acids or ester compounds thereof such as isophthalic acid, dimethylisophthalate, orthophthalic acid, diethylorthophthalate, naphthalene dicarboxylic acid, or dimethyl naphthalene dicarboxylate and polymer compounds such as 1,4-butane diol, neopentyl glycol, 1,4-cyclohexane dimethanol, and propylene glycol.

The polyester prepolymer pellets used in the invention are manufactured by blowing an inert gas into the polymer melt in order to carry out pelletization, with this process being carried out from 60 minutes prior to completion of the polycondensation until the reaction is completed, or more preferably, until the completion of polycondensation.

In this case, an inert gas such as nitrogen, argon, helium, or neon may be used, with nitrogen being the preferred substance. The volume of the inert gas blown in should be 0.1 to 5,000%, and preferably 0.5 to 300%, with respect to the volume of the polymer melt. An extrusion die for direct blowing and distribution of the inert gas may be installed for blowing the inert gas into the polyester prepolymer.

The polyester pellets containing the inert gas used in manufacturing according to the above method are then placed under a vacuum during the crystallization stage, thus molding them into chips having the appearance of popcorn. The degree of vacuum used is 10 torr or less.

In this case, if the above vacuum is greater than 10 torr, it will take too long to form the popcorn-shaped chips, producing an unfavorable result. A suitable temperature range is 75 to 230°C, and preferably 100 to 180°C. If the temperature is too low, the popcorn chips will not be formed, and if it is too high, although crystallization will take place, the formation of the popcorn-shaped chips will not occur.

It is important to note that the bubbles in the prepolymer must have popped before crystallization takes place. If crystallization is carried out before the bubbles inside the prepolymer have popped, it will have a shape identical with that of ordinary polyester prepolymers, and the popcorn-shaped chips will not form.

In manufacturing of the popcorn-shaped polyester resin by the above method, these polyester pellets will typically be heated for 5 to 300 minutes at 120 to 180°C and crystallized

with a target degree-of-crystallization of 20% or more. Adhesion of the polyester pellets is prevented in the drying process.

A dehumidifying dryer can be used to decrease the acetaldehyde content of the popcorn-shaped polyester resin crystallized as described above. In carrying out drying to remove acetaldehyde, the temperature is increased to 160 to 200°C, and the polyester resin is then passed through the dehumidifying dryer for 2 to 20 hours, and preferably 3 to 10 hours, making it possible to decrease its acetaldehyde content to 1 ppm or below. If the above drying, for the purpose of the removal of the acetaldehyde, is carried out at 160° or below, the acetaldehyde-removing effect will be small, and if the temperature exceeds 200°C, the color of the polyester resin will change and thermal decomposition and oxidative decomposition will occur. Moreover, the dew point of the dehumidified air used in this case should be -20°C or below, and preferably -40°C or below, and the volume used should be 0.05 to 1,000 L per kg of the polyester resin.

In addition to dehumidified air, the above acetaldehyde removal process may also be carried out using a vacuum or inert gas, with suitable inert gases including nitrogen, carbon dioxide, helium, argon, and neon. The intrinsic viscosity of the polyester resin according to the invention should be in the range of 0.6 to 1.3 dL/g in order to maintain the physical properties of the container.

Using the method of the invention, a polyester resin for use in food containers can be manufactured by an economical method that does not require the use of solid-state polymerization. In addition to polyester resin, the method can also be used for the removal of impurities such as volatile substances from all resins.

The following is an explanation of the invention in further detail by means of working examples, but the invention is by no means limited by these examples.

In Working Examples 1-6 and Comparison Examples 1-3 below, the intrinsic viscosity of the manufactured PET polymer pellets and PET resin was measured at 30°C using a mixed solvent composed of phenol/tetrachloroethane (in a ratio 6 : 4 by weight). Furthermore, in order to measure the acetaldehyde content of the PET polymer pellets and PET resin, 5 g of PET pellets or PET resin was frozen using liquid nitrogen and pulverized, 1.0 g of the pulverized sample was placed in a tube and heated for 10 minutes at 160°C, a 5 mL sample was removed from the tube, and this sample was measured by gas chromatography.

Working Example 1

5 minutes prior to the completion of the polycondensation of polyethylene terephthalate (PET) polymer, 5 vol. % of nitrogen with respect to the volume of PET polymer was injected via the bottom of the reactor, the mixture was stirred, and extrusion and cutting were carried out to

obtain round pellets measuring 3 x 3 x 2 mm. The intrinsic viscosity of the PET polymer pellets was 0.75 dL/g, and their acetaldehyde content was 46 ppm.

50 g of prepolymer chips containing round bubbles were placed in a rotatory vacuum distiller and heated under a vacuum at 120°C for 30 minutes. In order to convert the PET polymer pellets into a popcorn shape, the temperature was continuously increased to 140°C and crystallization was carried out for 60 minutes.

After this, in order to remove the acetaldehyde, the vacuum was released, the temperature was increased to 170°C, and air with a dew point of -42°C was passed through zeolite at a rate of 50 mL per minute for 4 hours. The acetaldehyde content of PET resin subjected to the above process was 1.3 ppm.

Working Example 2

The same method as in Working Example 1 was used, except that the temperature for the acetaldehyde removal was set at 170 to 180°C. The acetaldehyde content of PET resin subjected to this process was 0.9 ppm.

Working Example 3

The same method as in Working Example 1 was used, except that the temperature for the acetaldehyde removal was set at 170 to 180°C. The acetaldehyde content of PET resin subjected to this process was 0.8 ppm.

Working Example 4

The same method as in Working Example 1 was used, except that during drying, for the purpose of the removal of the acetaldehyde, the dehumidified air was passed through for 6 hours rather than 4 hours. The acetaldehyde content of PET resin subjected to this process was 1.0 ppm.

Working Example 5

The same method as in Working Example 2 was used, except that during drying, for the purpose of the removal of the acetaldehyde, the dehumidified air was passed through for 6 hours rather than 4 hours. The acetaldehyde content of PET resin subjected to this process was 0.8 ppm.

Working Example 6

The same method as in Working Example 3 was used, except that during drying, for the purpose of the removal of the acetaldehyde, the dehumidified air was passed through for 6 hours rather than 4 hours. The acetaldehyde content of PET resin subjected to this process was 0.5 ppm.

Comparison Example 1

PET resin pellets were manufactured by means of the usual method without injecting inert gases during polycondensation of the PET polymer. The intrinsic viscosity of the PET was 0.76 dL/g. These PET resin pellets were placed in the same rotatory vacuum distiller used in Working Example 1, and while heating to 120°C, crystallization was carried out for 30 minutes under a vacuum. After this, the vacuum was released, the temperature was increased to 170°C, and air having a dew point of -42°C was passed through at a rate of 50 mL per minute for 4 hours. The acetaldehyde content of PET subjected to this process was 1.85 ppm.

Comparison Example 2

The same method as in Comparison Example 1 was used, except that the temperature during acetaldehyde removal was set at 170 to 180°C. The acetaldehyde content of PET resin subjected to this process was 14.1 ppm.

Comparison Example 3

The same method as in Comparison Example 1 was used, except that the temperature during acetaldehyde removal was set at 170 to 180°C. The acetaldehyde content of PET resin subjected to this process was 11.3 ppm.

(57) Claims

Claim 1

A method for manufacturing a polyester resin for use in food containers, characterized in that, in the process of manufacturing the polyester polymer, an inert gas is blown into the polymer melt to form polyester prepolymer pellets containing air bubbles, and while these are heated at 100-180°C, a vacuum of 10 torr or less is applied to form the polymer pellets into a popcorn shape, after which crystallization and drying are carried out.

Claim 2

The method for manufacturing a polyester resin for use in food containers according to Claim 1, characterized in that the aforementioned polyester prepolymer contains polyethylene terephthalate or 80 mol % or more of ethylene terephthalate structural units.

Claim 3

The method for manufacturing a polyester resin for use in food containers according to Claim 1, characterized in that the above drying is carried out at a temperature of 160-200°C.